The effect of the individual species of the N plasma on the characteristics of InAsN quantum dots grown by MBE

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Abstract

The influence of the different species which constitute N plasma, such as atomic nitrogen, diatomic nitrogen and ionized species, on the morphological and optical characteristics of the InAsN quantum dots (QDs) has been studied in this work. We have performed several sets of growths modifying in each one the concentration of these species. Atomic force microscopy (AFM) and photoluminescence (PL) techniques have been used to perform the surface characterization and the optical analysis of these samples, respectively. Clearly, we have found a strong correlation between the structural and optical characteristics of the InAsN QDs with the plasma composition used during the growth. Ionized species favour the high density of QDs, atomic nitrogen increase dimensions of the QDs and molecular nitrogen does not almost affect the characteristics of these nanostructures. An increment of ionized species in the plasma yields a higher density of QDs, an increase in the atomic nitrogen increases the dimensions of the QDs and the molecular nitrogen flux used does not almost affect the characteristics of these nanostructures. Also, we have found that there is not redshift of the peak wavelength of the PL emission as we increase the atomic nitrogen concentration during the growth. This may be due to equal nitrogen incorporation into the quantum dots. We supposed that the mechanism dominating in it is possible that the nitrogen incorporation in these types of nanostructures depend on another growth parameter.

1. Introduction

It has been recently demonstrated that In(Ga)As quantum dots (QDs) are very attractive nanostructures for optoelectronic devices fabrication in fiber communications systems in 1.3 μm and 1.55 μm, due to their special and unique physical properties such as the three dimensional carrier confinement However they have some limitations related to the extension of the wavelength emission. When we increase too much the sizes of the QDs dislocations could appear due to the huge strain accumulated in these nanostructures. So, in order to avoid these inconveniences, another possible way to extend the wavelength emission is the incorporation of N into the In(Ga)As QDs

As it is usual in the molecular beam epitaxy (MBE), the nitrogen source used to supply active nitrogen into the material grown is a radiofrequency (RF) N plasma source. Nevertheless, this source not only supplies the active (atomic) nitrogen, species that incorporate in the material grown, but several other species coexist in this plasma, for instance, electrons, diatomic nitrogen and ionized species. Therefore, it will be very interesting to study the influence of these different species of the nitrogen plasma in the characteristics of the In(Ga)AsN QDs. In this work the main objective has been to study the effect of the individual species that constitute the N plasma in the morphological (size, height and density) and optical (peak wavelength of the photoluminescence (PL) emission, PL intensity, etc.) characteristics of the InAsN QDs grown by MBE on GaAs (1 0 0) surfaces. In this way, several InAsN QDs samples were grown modifying the concentration of the individual species of the N plasma. Atomic force microscope (AFM) is used to characterize the uncapped QDs, whilst PL measurements are used to investigate their optical properties when capped with GaAs. The results show that the characteristics of the InAsN QDs depend clearly on atomic nitrogen and ionized species concentrations. However, if we modify the molecular nitrogen concentration, the QDs characteristics hardly are affected.

2. Experimental procedure

The samples were grown on semi-insulating GaAs (1 0 0) by a RIBER 32, solid source MBE equipped with a RF Oxford Applied Source which supplies the atomic nitrogen. The structure of these samples was the following: a 500 nm GaAs buffer layer was grown on a GaAs substrate at 590 °C and a rate of 1 μm/s, and then the substrate temperature was dropped to 450 °C for the growth of the InAsN QDs. The equivalent amount of material and the growth rate were 4 ML and 0.15 ML/s, respectively, and the V/III flux ratio was 40. According to the experience in our group in the growth of dilute nitrides, it is strongly recommended to avoid the nitridation of the surface, since structural and optical quality of the resulting material could get damaged. Therefore, we have followed a growth method to avoid the nitridation of the surface on which later we have grown the InAsN QDs. This method consists of three fundamental steps: (1) Just before at the beginning of the growth, the main shutter is closed; (2) then N plasma source is switched on; and (3) QDs growth is carried out, so nitrogen, In and finally the main shutter are opened The N plasma source conditions (nitrogen flux and plasma power) were set in each experiment depending on the species that we were studying in each experiment, as detailed below. A 50 nm GaAs layer grown at 450 °C were used to cover the QDs. Later, the substrate temperature was increased to 590 °C, and a 250 nm GaAs layer was grown to bury the QDs and flatten the surface, as observed *in situ* by reflection high energy electron diffraction (RHEED). Finally, on the surface of the sample an additional layer of InAsN QDs was grown under the same growth conditions as the buried QDs, to make the structural characterization of these nanostructures using AFM operating in tapping mode. Buried QDs were used to perform the optical characterization at low temperature (15 K). For the PL measurements, the samples were mounted in a closed-cycle helium cryostat and excited using a 781 nm laser. A liquid nitrogen-cooled germanium detector was used to detect the emission using a conventional lock-in technique.

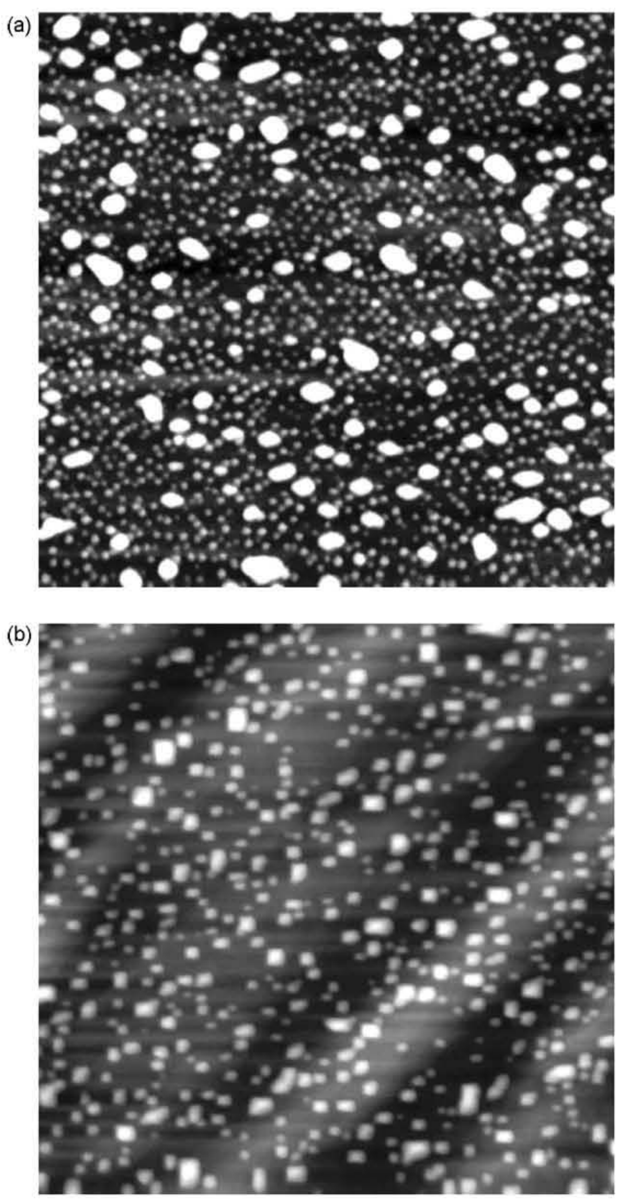

Fig. 1. AFM images (1 μm × 1 μm). (a) InAs QDs (height scale of 6 nm) and (b) InAsN QDs (height scale of 35 nm) grown under the same growths conditions on GaAs (1 0 0) substrates.

The first of our experiments was to study the influence of the N plasma with all species at the same time on the growth of InAsN QDs. So, we made a comparison between the InAs and the InAsN QDs, both growing under the same growth conditions. And later, we carried out three sets of growths and in each one we have analysed the influence of the individual species of N plasma (ionized species, atomic nitrogen and molecular nitrogen).

3. Results and discussions

The effects of the nitrogen incorporation into the structural properties of InAsN QDs were assessed through the analysis

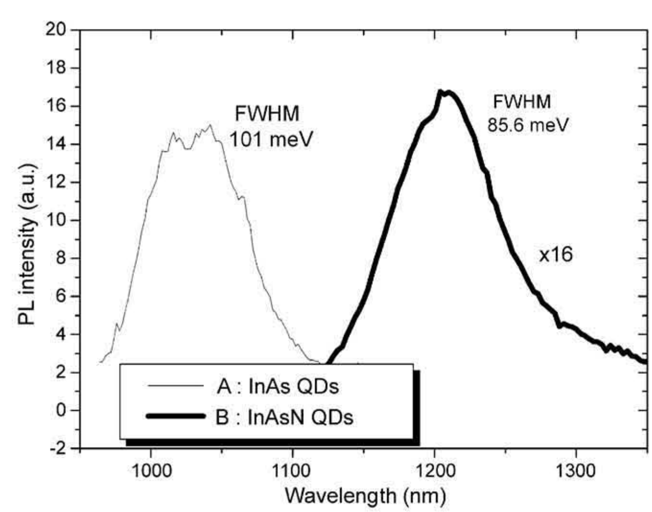

Fig. 2. Low temperature (15 K) PL measurements of InAs and InAsN QDs grown under the same growth conditions (their morphological characteristics are shown in Fig. 1).

of the AFM images shown in Fig. 1. A clear increase of the dimensions and a reduction of the InAsN QDs density compared to those of InAs QDs are to be seen. We supposed that one of the reasons of these results could be the reduction of the lattice mismatch as N incorporates into InAs QDs. InAsN grown on GaAs shows higher critical layer thickness than the one of InAs. This is consistent with the different observed formation times for the InAs and InAsN nanostructures (based on RHEED pattern observations), which were longer for the dilute nitride QDs. Hence, when the InAsN QDs are formed their dimensions are larger and density is lower. The height and the density of the InAsN QDs grown is about 12–15 nm and 5×10^{10} QDs cm^{-2} approximately, respectively, and in the case of InAs QDs grown under the same growth conditions the density is about 2.5×10^{11} QDs cm^{-2}, and two distributions of sizes have been found: 4.3 nm and 10.5 nm of height. According to the results of the PL measurements (Fig. 2), the peak wavelength of the PL of the QDs emission is redshifted and the PL intensity is weaker than that measured in the InAs QDs sample, as expected. As occurred in InGaAsN quantum wells the optical quality of the QDs could be degraded due to the addition of nitrogen into the QDs. Some of the reasons could be the formation of point defects and the damage induced by the ions.

In Fig. 3, we can observe the AFM images and the PL measurements of the first set of samples grown to study the influence of the individual species of the N plasma. This set was oriented to study the effects of the ionized species in the InAsN QDs. The experiment consisted of the growth of three equivalent samples in which the only difference among them was the ions concentration in the plasma used for their growth. This was done by

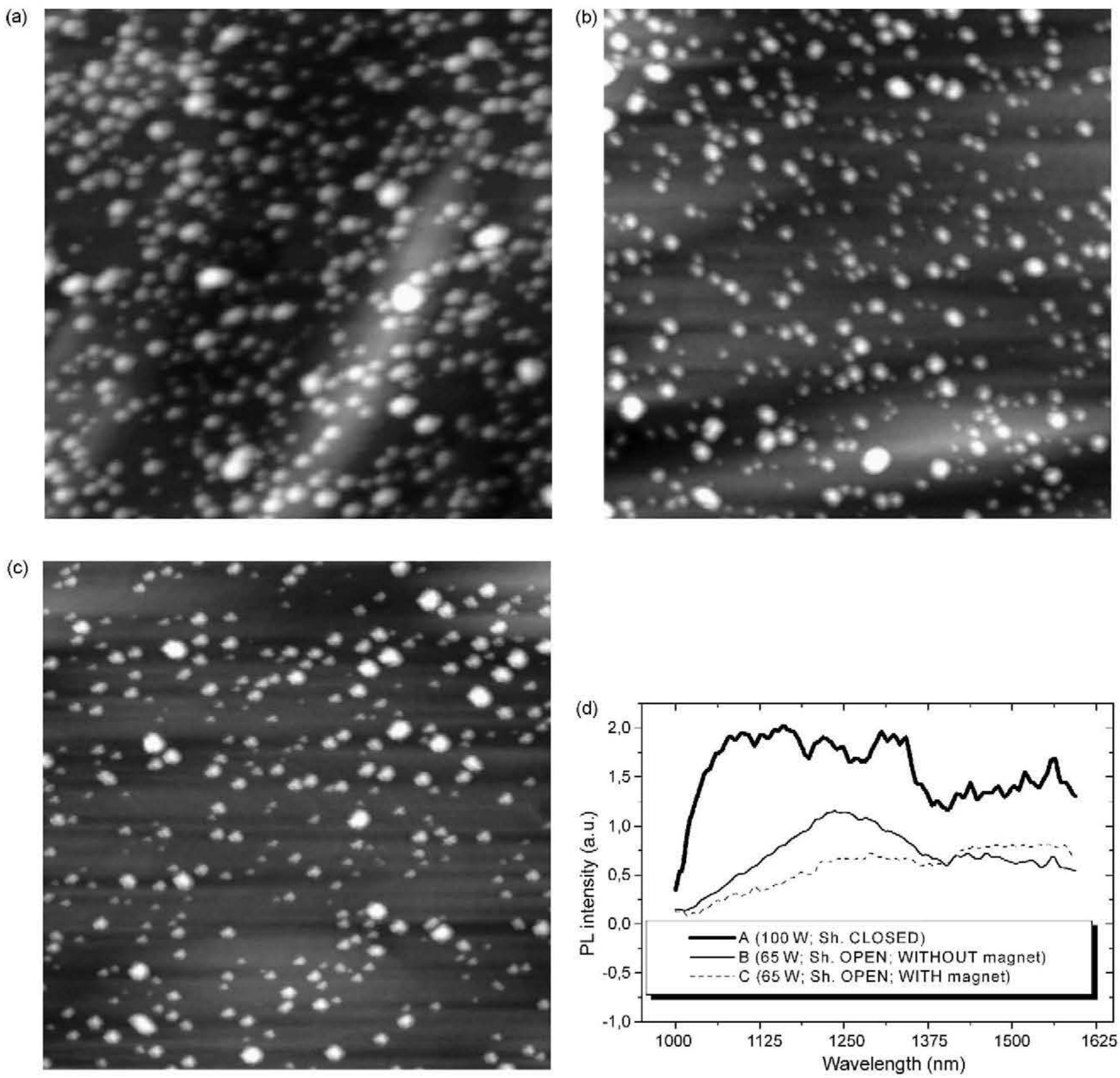

Fig. 3. AFM images (1 µm × 1 µm) with a height scale of 30 nm (a–c) and low temperature (15 K) PL measurements (d) of InAsN QDs grown at 0.15 ML/s with different ions concentration using different N plasma source configurations: (a) 65 W, (b) 65 W + magnetic field and (c)100 W + N shutter closed.

changing the amount of charged species impinging onto the sample by changing the plasma power and magnetic deflection using a 0.2 T static magnet. Two of these samples were grown using the same plasma power (65 W), although in one of them a magnetic field was applied in order to deflect the ions and to avoid these ions reach to the surface of the sample (lower ions concentration). A third sample was grown using a higher plasma power (100 W) to get a higher ions concentration during the growth. Since the N_2 flux was fixed in the three samples at 0.2 sccm, the atomic nitrogen concentration in this third sample should be larger than in the other two cases, so to get the same active N concentration we closed the N shutter for this last sample. The structural results indicate that when we increase the ions concentration in the plasma, the density of QDs tends to increase (from 4.4×10^{10} QDs cm^{-2} (Fig. 3(c)) to 6.4×10^{10} QDs cm^{-2} (Fig. 3(a))) and larger QDs appear (from a height and a width of 4 nm and 20 nm, respectively (Fig. 3(c)) to 11 nm and 42 nm, respectively (Fig. 3(a))). The results of the PL measurements of these samples are presented in Fig. 3(d). It should be noticed here that the PL intensity is correlated with the morphological results. Additionally, we made another experiment which may allow us to clarify the influence of the ions on InAsN QDs. In this case, samples with the same structure were grown but using a slower growth rate of 0.07 ML/s. In Fig. 4, we can observe that the higher ions concentration increases the density and the size of InAsN QDs. Indeed, once more, PL intensity is correlated to the morphological results.

Then we carried out several growths to study how the diatomic nitrogen affects the characteristics of the InAsN QDs. When we grow using N plasma, the background pressure of the growth chamber increases between two and three orders of magnitude because of the fact that the shutters of the cells are not

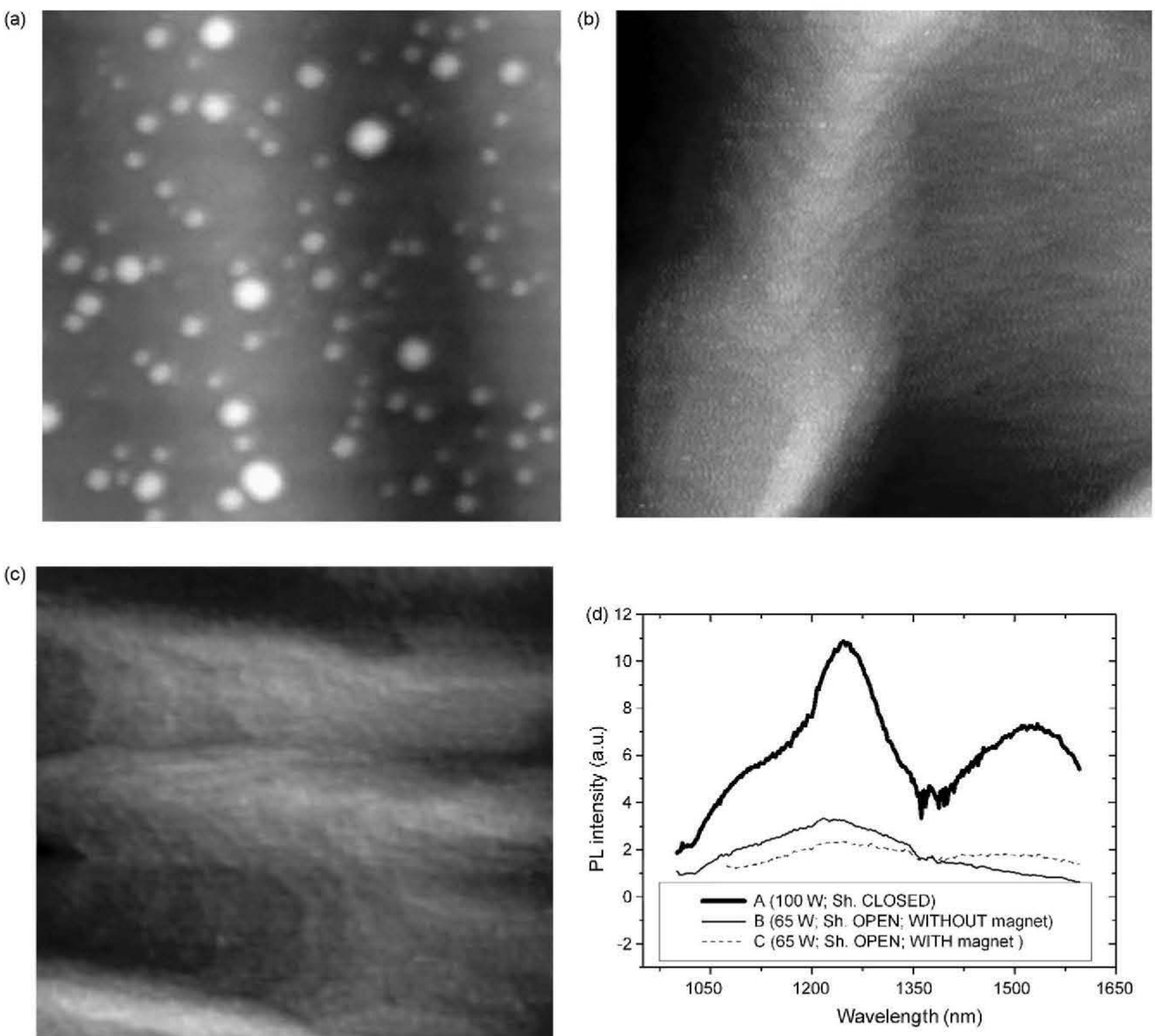

Fig. 4. AFM images (500 nm × 500 nm) with a height scale of (a) 25 nm, (b) 9 nm and (c) 7 nm, and low temperature (15 K) PL measurements (d) of InAsN QDs grown at 0.07 ML/s with different ions concentration using different N plasma source configurations: (a) 65 W, (b) 65 W + magnetic field and (c) 100 W + N shutter closed.

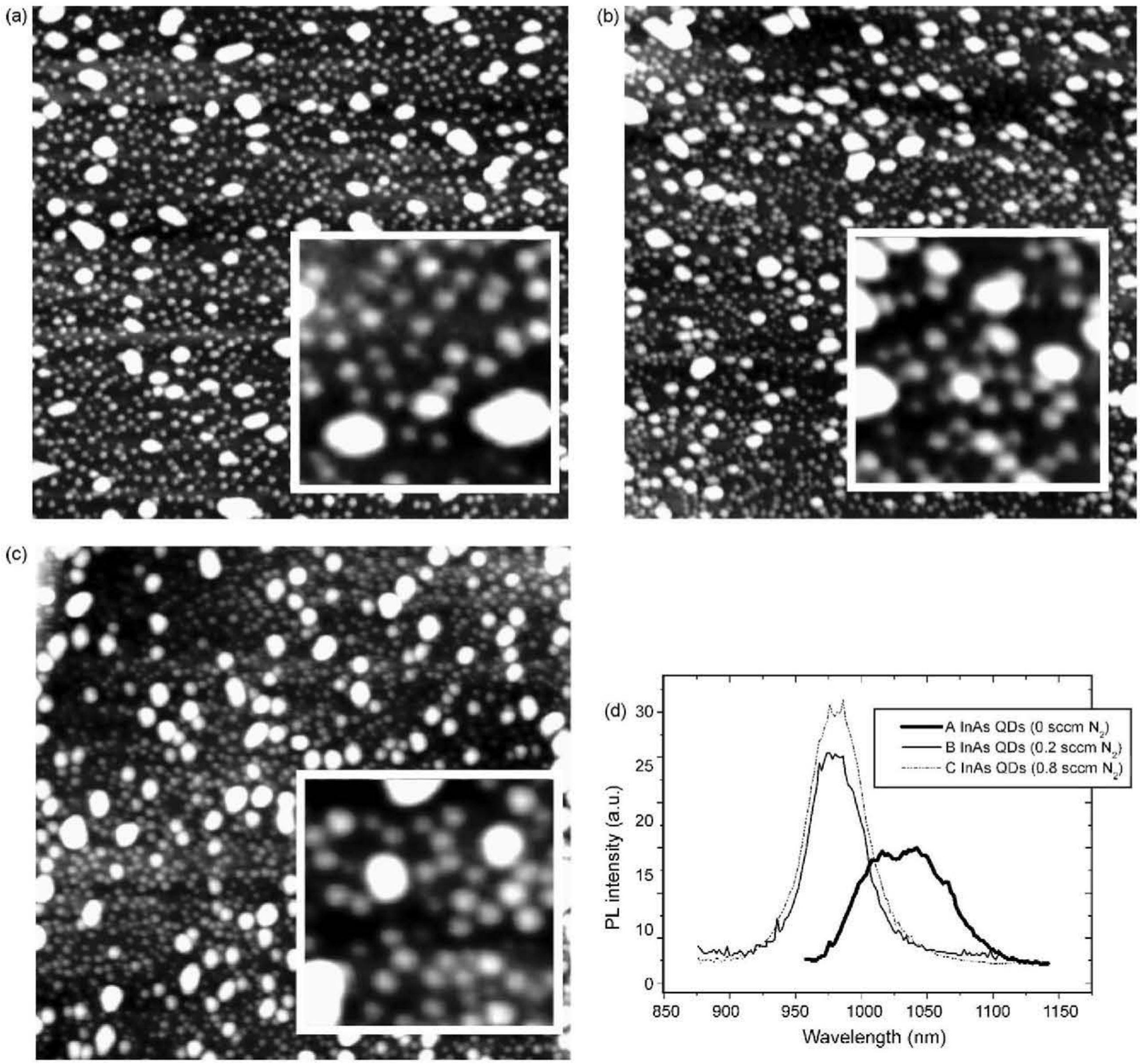

Fig. 5. AFM images (1 μm × 1 μm) with a height scale of 6.3 nm (a–c) and low temperature (15 K) PL measurements (d) of InAs QDs grown under different N_2 fluxes: (a) 0 sccm, (b) 0.2 sccm and (c) 0.8 sccm.

hermetically sealed. Hence, the pressure of the growth chamber increases too much when we generate the N plasma using a N_2 gas. This study has shown how the high pressure of the growth chamber affects the characteristics of the InAsN QDs. We have grown a set of three similar samples: (1) without a N_2 flux (Fig. 5(a)), (2) with a N_2 flux of 0.2 sccm (Fig. 5(b)) and (3) with a N_2 flux of 0.8 sccm (Fig. 5(c)). We decided to grow InAs QDs instead of InAsN QDs to keep the other species concentrations fixed easily, i.e. ions and atomic nitrogen concentration were absent. With regard to the analysis of the AFM images of the surface of these samples, we found that the density of the QDs is approximately the same in all the three cases (3×10^{11} QDs cm^{-2}). Nevertheless, the presence of molecular nitrogen during the growth introduced an almost negligible increase of the density of QDs, and a slight reduction of the height of these nanostructures (a difference of ~1 nm). PL measurements of these samples seem to be agreeing with the morphological results (Fig. 5(d)). When the growth is under an ambient of molecular nitrogen, there is a slight improvement of the PL intensity and a weak blueshift of the peak wavelength of the PL emission. But, there is almost no significant differences between the results obtained growing under 0.2 sccm and 0.8 sccm of N_2 flux. In consequence, we can conclude that the molecular nitrogen and the presence of a high pressure N_2 atmosphere hardly affect the characteristics of InAsN QDs.

Finally, we will discuss the role of the atomic nitrogen of the N plasma in the InAsN QDs. Three equivalent growths were performed in which the only growth parameter changed was the plasma power, to achieve different atomic nitrogen concentration in the plasma. The nitrogen flux was adjusted to 0.2 sccm and the amount of active nitrogen generated was monitorized during growth by an optical emission detector (OED). More-

Table 1
Size and height of the InAsN QDs for a set of samples grown with different atomic nitrogen concentrations

OED	Height		Size	
	Smaller QDs (nm)	Larger QDs (nm)	Smaller QDs (nm)	Larger QDs (nm)
30	8	15	40	47
58	15	23	67	90
85	23	32	75	146

Two different distributions of QDs are shown (smaller and larger QDs).

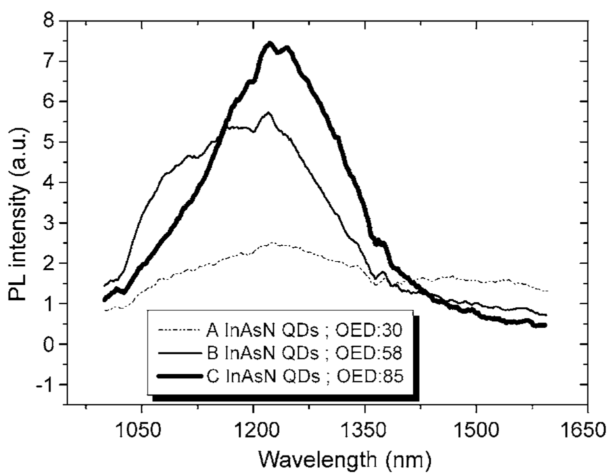

Fig. 6. Low temperature (15 K) PL measurements of the set of InAsN QDs grown with different atomic N concentration using different plasma powers: (a) 55 W (OED: 30), (b) 65 W (OED: 58) and (c) 85 W (OED: 85).

over, a magnetic field (0.2 T) was applied in all of them to get the same ions concentration in all of them. AFM measurements of these samples suggest that the introduction of higher atomic nitrogen concentration gives rise to an increase of the dimensions of the QDs (Table 1), a greater density of larger QDs and a reduction of smaller QDs density. As we can observe in the optical measurements in Fig. 6, an improvement of the PL intensity is to be seen as the amount of atomic nitrogen is increased. However, there seems not to be a redshift of the peak wavelength of the PL emission as one would expect if the active nitrogen concentration increases. Thus, the nitrogen incorporation into the QDs may be equal in all the three growths. So, we can conclude temptatively that the nitrogen incorporation into the QDs may depend more strongly on other growth parameters. Currently, further work is in progress to clarify these results.

4. Conclusions

In this work, we have studied the influence of the different species that constitute the N plasma on the morphological and optical characteristics of InAsN QDs. We found that ionized species of the plasma increased the density of the InAsN QDs and the formation of larger QDs. We could additionally conclude that molecular nitrogen hardly affects in the characteristics of the InAsN QDs. Additionally, the density of the larger InAsN QDs tends to increase under a high atomic nitrogen concentration growth conditions. Nevertheless, peak wavelength of the PL emission was not redshifted in these preliminary results when active nitrogen was increased, using the above describe conditions. It is likely that the nitrogen incorporation depends also on another growth parameter.

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